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PRELIMINARY REPORT ON THE RELEASE OF FISSION PRODUCTS ON MELTING GE-ANP FUEL

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1.0 Summary.

The initial experiments in the melting of GE-ANP ceramic fuel tubes by the carbon arc-image process have been completed. The method besides easily reaching the required temperature has many advantages of both experimental and analytical nature. Its chief limitation lies in that all of the heating is produced on the surface by radiation and thereby implies a significant gradient for short heating periods. In melting a ceramic specimen, surface temperatures may possibly exceed the average temperature by several hundred degrees. This effect is being investigated.

Results of the first series of the experiments with the uncoated small cylindrical type fuel (LTC-14, 0.43% burnup) gave an average release of about 10 percent of the gross gamma activity, 73 percent of the gases, 70 percent each of the iodine and tellurium, 50 percent of the cesium and 60 percent of the ruthenium. With an airflow of less than 1 foot per minute across the sample, approximately 90 percent of all the fission products (except the rare gases) plated out on the cold furnace tube in a coating of vaporized uranium and beryllium oxides. At an air flow of several feet per minute, this effect decreased to about 30 percent and most of the activity and metallic oxides passed through the furnace to the microfilters. Iodine diffused on past the filters to the special hot charcoal trap designed to retain it. Of the remaining fission products: the rare earths; the alkaline earths (Ba and Sr) and the refractory metals, less than 0.5% was released from the melt. The strontium value was approximately 0.15%. About 8 percent of the uranium was vaporized along with 1 percent of the BeO.

Trace concentration of fission products (0.01% burnup) gave a somewhat diminished release rate compared to that observed after 0.43% burnup.

Experiments are continuing to include two types of coated fuel.

2.0 Abstract

A short-term program was undertaken to furnish preliminary data for the evaluation of the hazards of radioactive fission-product release in the event of a melt-down in the GE-ANP type reactor. The method of optically focussing radiant heat from a carbon arc has been used. In this process no container crucible is used since the melted portion is retained by the remainder of the sample. Of three types of fuel to be tested, results are presented on only the first.

3.0 Introduction

The ANP direct-cycle ceramic fuel is a tubular extrusion of stabilized BeO-5% UO_2 which is designed to withstand temperatures beyond those practical with metal systems. The fact that structural integrity is maintained even at temperatures approaching the melting point suggests advantages in terms of power production but allows a lesser margin between operation and accidental melting.

In order to appraise such a maximum accident it is of greatest importance to have available an acceptable set of values for the release behavior of individual fission products. For metallic systems fractional releases are generally proportional to melting temperature and can be estimated according to type from results on similar materials. See ORNL-2616.

To extend such data to both a non-metallic (ceramic) system and to extrapolate to an abnormally high temperature seemed to have to much uncertainty to merit consideration. For this reason it was felt necessary to obtain experimental results by as realistic a method as possible. Since a program was already at the point of procuring similar values for the widely developed civilian power reactor fuel, UO_2, a temporary diversion was agreed upon to explore BeO-UO_2. As a logical step up the temperature scale, this mixture provided an introduction to the problems of attaining these high temperatures.

4.0 Experimental Techniques Available

In order to melt BeO-5% UO_2, a temperature of approximately 2450° C is necessary. As a further complication of the experimental requirements, the presence of a high velocity air stream narrowed the selection of a method to the following possibilities.

(1) Electric-Arc Melting, by a non-consumable electrode process. This would imply an adaptation of the vacuum-arc or inert gas-arc process which in air leads to extensive sputtering and copious evaporation of UO_3. Difficult reclamation of material fused into container materials added to the undesirability of this process.

(2) RF-Induction Melting, based on contact with a susceptor additive such as stainless steel. It had been demonstrated that by heating molten stainless steel to the boiling temperature, UO_2 could be melted in a ZrO_2 crucible. The ZrO_2 acts as a diluent in solid solution of UO_2. Unfortunately, the effect of both the metallic susceptor and the diluent are unknown factors as yet to be evaluated.

(3) The Carbon Arc-Image Furance. This method had been previously evaluated on test samples using a vendors demonstrator machine[1]. The technique has also been well characterized by work at National Carbon Co.[2]. Because of its assurance of attaining the temperature and its freedom from container problems as well as contact with secondary materials, this method was chosen.

4.1 Experimental Arrangement for Melting $BeO-UO_2$

The GE-ANP hollow tubular type fuel is found to be ideally suited to the limitations of the Arc-Image furnace. The diameter of the image in which a sufficiently high heat flux is available to melt BeO is little more than one centimeter. ANP fuel tubes range from 0.7 to 1.1 centimeters with a center hole of 0.4 to 0.7 cm. While the peak temperature in the image may exceed that required for melting, experimental conditons allow for the melted portion of the sample to deflect out of the high temperature zone in a downward direction and to present a cooler portion to the impinging beam.

Containment is one of the principal advantages of this type furnace because of the ease of transmission of the radiant heat through the walls of ordinary laboratory glass ware. No special arrangements are necessary and maximum freedom is possible in setting up the sample. At the temperatures anticipated a significant vaporization of uranium and perhaps beryllium as oxides was anticipated therefore a completely closed system was constructed. Figures 1, 2, and 3 show the physical appearance of the furnace both before and after the addition of lead shielding. Figure 4 is a schematic outline of the fission product containment and collection train in relation to the arc furnace. The furnace tube is made mobile by mounting on the sample positioner which is electrically powered for movement in three co-ordinate axes. In this present work the furnace has been operated only in a horizontal position for ease in shielding. For some purposes vertical operation is required. A front reflecting 45° vycor mirror with water cooling must be installe for this position.

5.0 Operational Procedure

At the 300 ampere rating, the maximum heating time is that required to consume a 27" electrode which is about 6 minutes. If the specimen has been previously set at the focal point, to avoid delay in locating the maximum heating position, significant loss in this heating time need not be experienced. In working with highly irradiated sources it is even more advantageous to have the focus preset on a dummy sample. The appearance of a trial sample in the furnace tube is shown in Fig. 5. The 1" long x 1/4" specimen is mounted on a piece of pure sintered BeO rod on which a short length is ground to match the inside diameter of the UO_2 piece. The BeO rod in turn is supported in a quartz tube sealed into a standard taper fitting. In practice, with all of the collection train ready for use, an irradiated specimen is placed in the tube fitting remotely and then the fitting is pressed into the matching joint with a suitably long pair of tongs.

At this point the selected rate of air flow is initiated upward through the furnace tube across the sample and through the following series in order:

1. The pyrex furnace tube and ~ 20 inches of glass tubing.

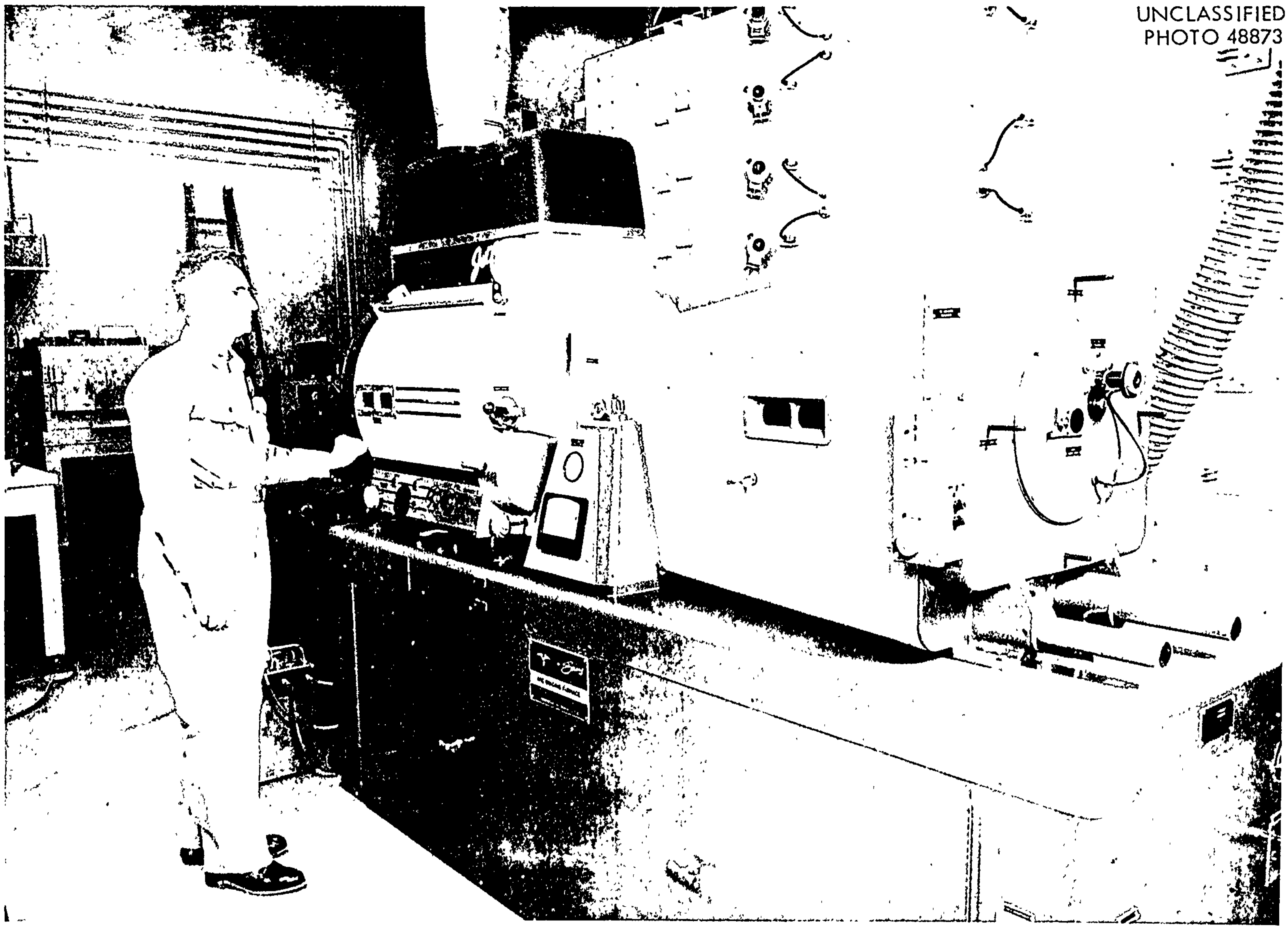

Figure 1
ARC IMAGE FURNACE IN INITIAL TRIAL OPERATION

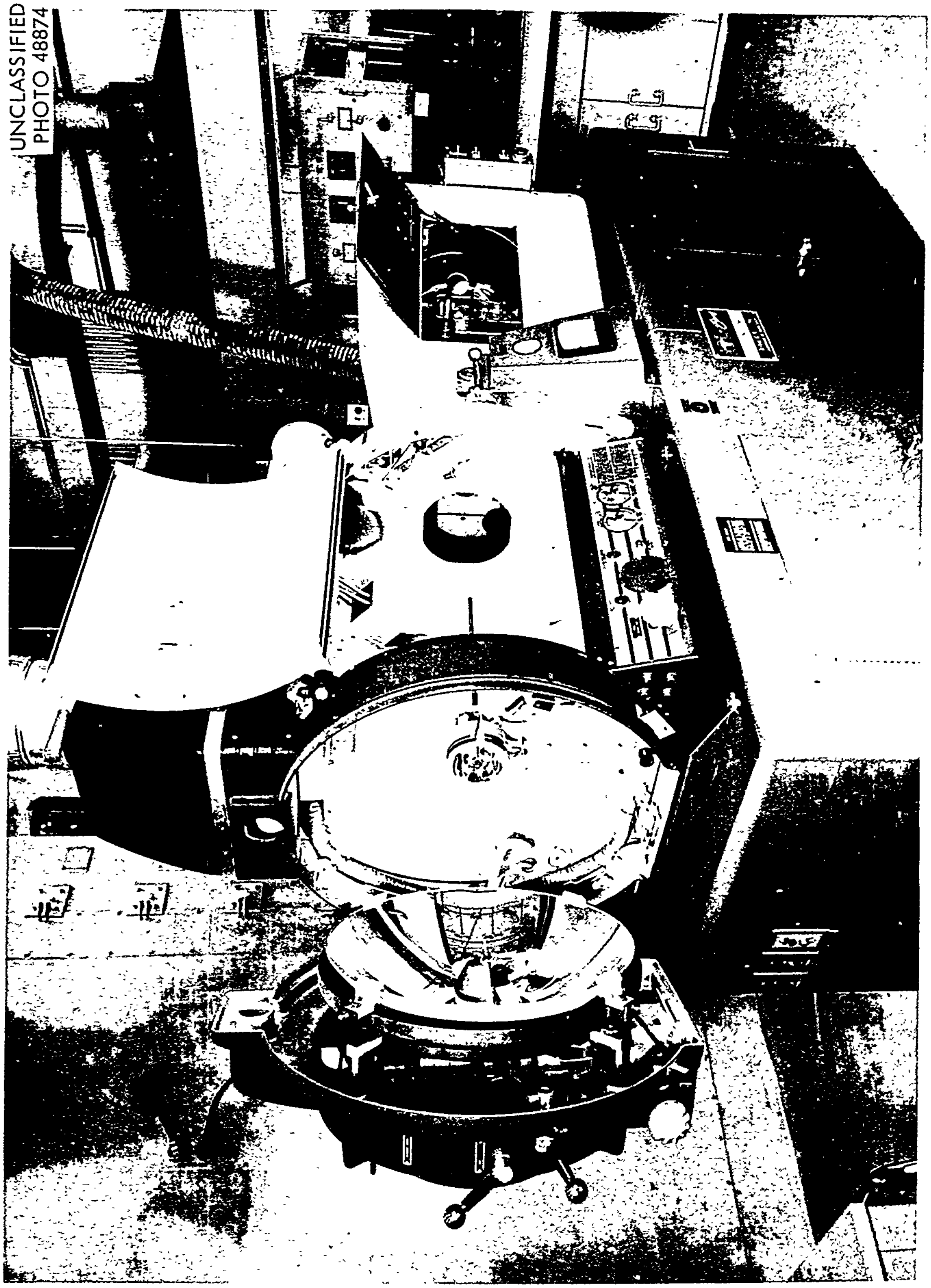

Figure 2
STRONG - ADL 300 AMP ARC-IMAGE FURNACE WITH SAMPLE TUBE
AS INSTALLED AT ORNL

Figure 3

VIEW OF ARC FURNACE TUBE AND ABSORPTION TRAIN

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1. ROTAMETER
2. MANOMETER
3. CHARCOAL TRAP - 195° C
4. COLD TRAP - 80° C
5. CHARCOAL TRAP 225° C
6. THERMOCOUPLE
7. MILLIPORE FILTER HOLDER
8. SAMPLE
9. RE-IMAGING MIRROR
10. VIEWING SHUTTER
11. SMOKED GLASS VIEWING PORT
12. OPTICAL PYROMETER
13. LEAD SHIELD (3 SIDES)
14. SAMPLE POSITIONER
15. FOCUS & ALIGNMENT FILTER
16. FIELD STOP
17. CHOPPING SHUTTER
18. DOUSER
19. ANODE
20. PROJECTION MIRROR
21. CATHODE
22. FURNACE TUBE

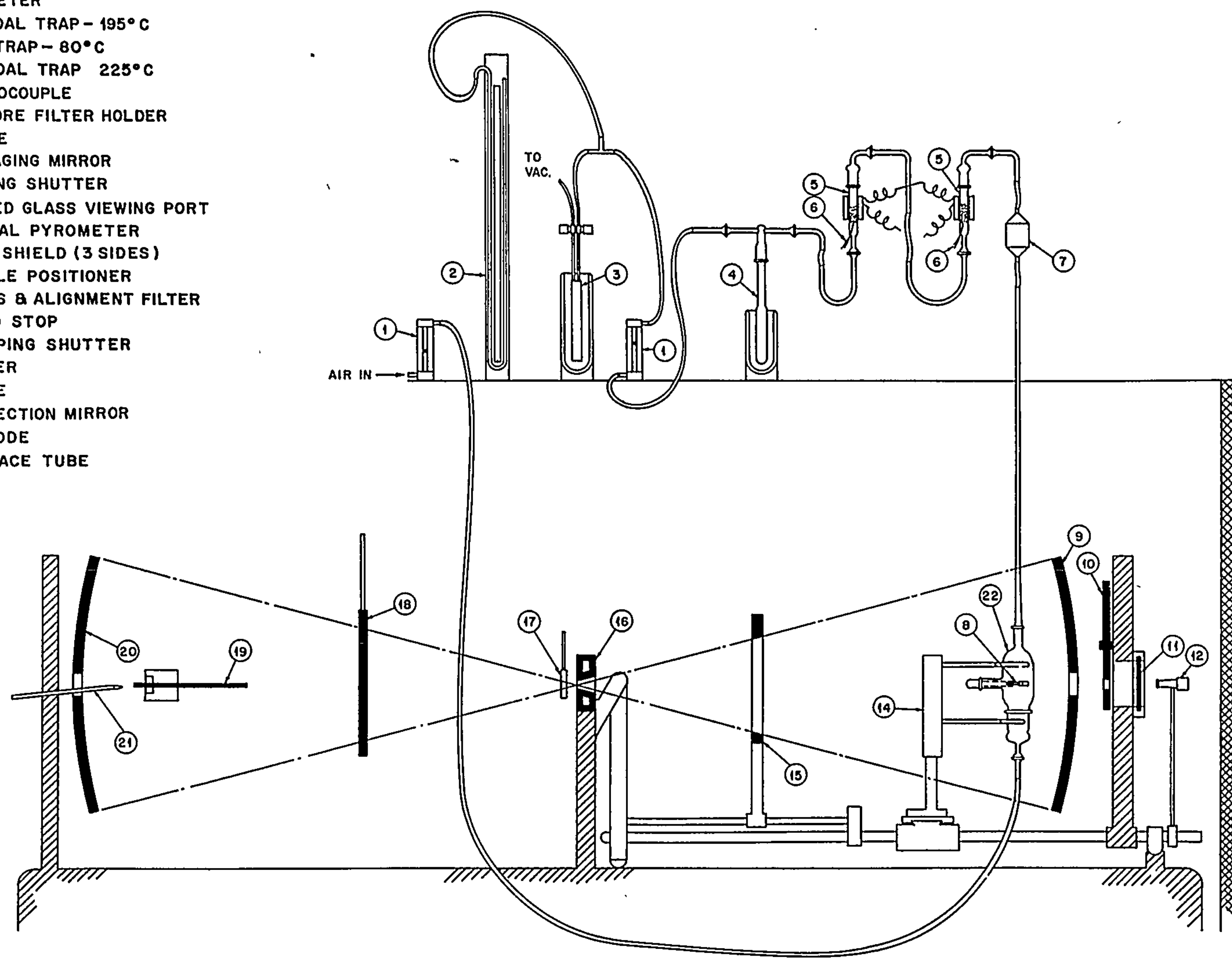

Figure 4
ARC-IMAGE FURNACE AND COLLECTION TRAIN
FOR FISSION PRODUCT RELEASE EXPERIMENTS

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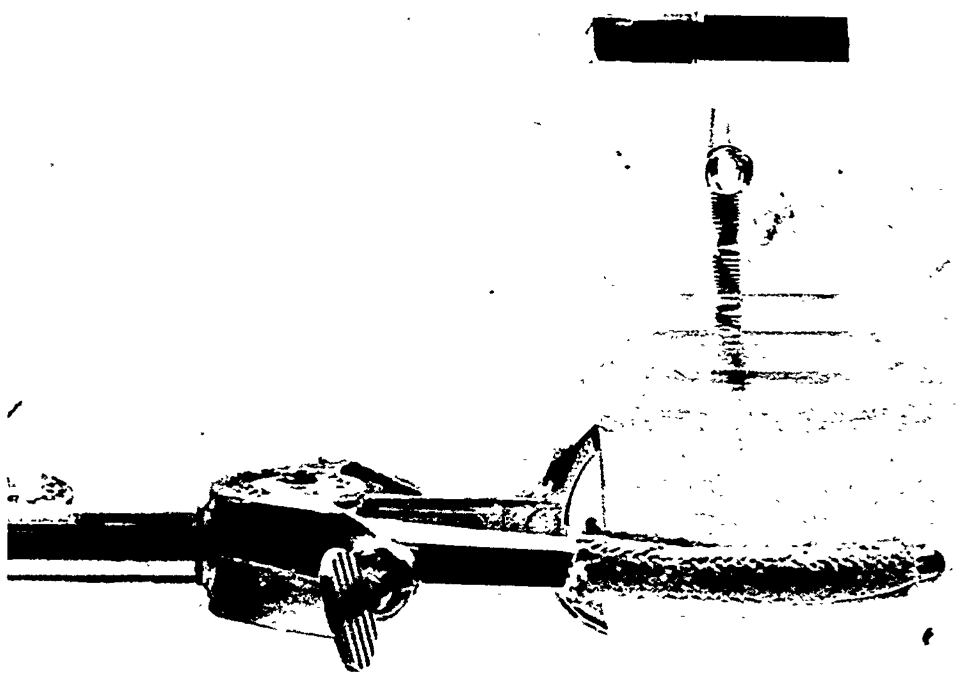

Figure 5

CLOSE-UP OF FURNACE TUBE SHOWING SAMPLE OF TYPE 14 FUEL IN PLACE

2. A stainless steel holder containing 2 - 47 mm. "millipore" filter papers of 3.5 microns and 0.8 micron mean pore diameter which collect all vaporized fission product solids. Only the rare gases and some iodine are collected beyond the filters.

3. Two iodine traps consisting of 10 ml. of activated coconut charcoal maintained at $\sim 225^{\circ}$ C to allow xenon and krypton to pass through.

4. A cold trap at -80° which removes only a small amount of water vapor.

5. A liquid nitrogen cooled charcoal trap containing about 50 ml of activated charcoal which adsorbs both the xenon and krypton.

The air flow having been adjusted throughout the system, the arc is initiated, stabilized, and the douser opened to start the heating cycle. By observation through the rear viewing port alignment is ascertained by relative brightness of a predetermined area. Within a few seconds melting on the front edge of the tube will have occurred. The sample appearance after heating may be like any of those in Fig. 6 but most often it will curve downward at 90° as in Fig. 7.

6.0 Heating Time and Temperature

At the present time, no accurate temperature measurements are being made since time has not permitted calibration of the optical pyrometer through the shutter mechanism. It is being assumed, for the present, that by the process of continuously advancing the specimen through the focal point, a length of less than 1 cm. is being heated at any given time. This implies that perhaps 1/3 of the specimen is molten at one time when the total heating time may be 90 seconds. It is further assumed that excessive temperatures are avoided by having the sample arranged to flow freely with gravity out of the heat zone as soon as melting occurs.

Time-at-temperature experiments have been considered with a smaller specimen mounted in such a manner that its size and proportions would permit complete sustained melting for a variable length of time. From such a series an effect of total time in the molten state could be assigned.

7.0 Vaporization of Uranium Oxide and Plate-Out on the Furnace Tube

The temperature of melting BeO is perhaps not sufficient according to the literature to volatilize UO_2 except in vacuo. However, in the presence of air volatilization of several percent of the uranium (probably UO_2 and UO_3) is observed as evident from Fig. 8. The material balances (Table I) show that several milligrams of the total ($\sim$ 86 mg. U^{235} per sample) are collected on the furnace tube and on the millipore filters. BeO is also volatilized to about 1/2 the rate of uranium. Curiously, when pure BeO is melted alone, no visible vaporization occurs. The filter papers plated with the vaporized oxides are a yellowish brown, suggesting UO_3. Typical papers are seen in Figs. 9 and 10. Particle size measurements which are in progress

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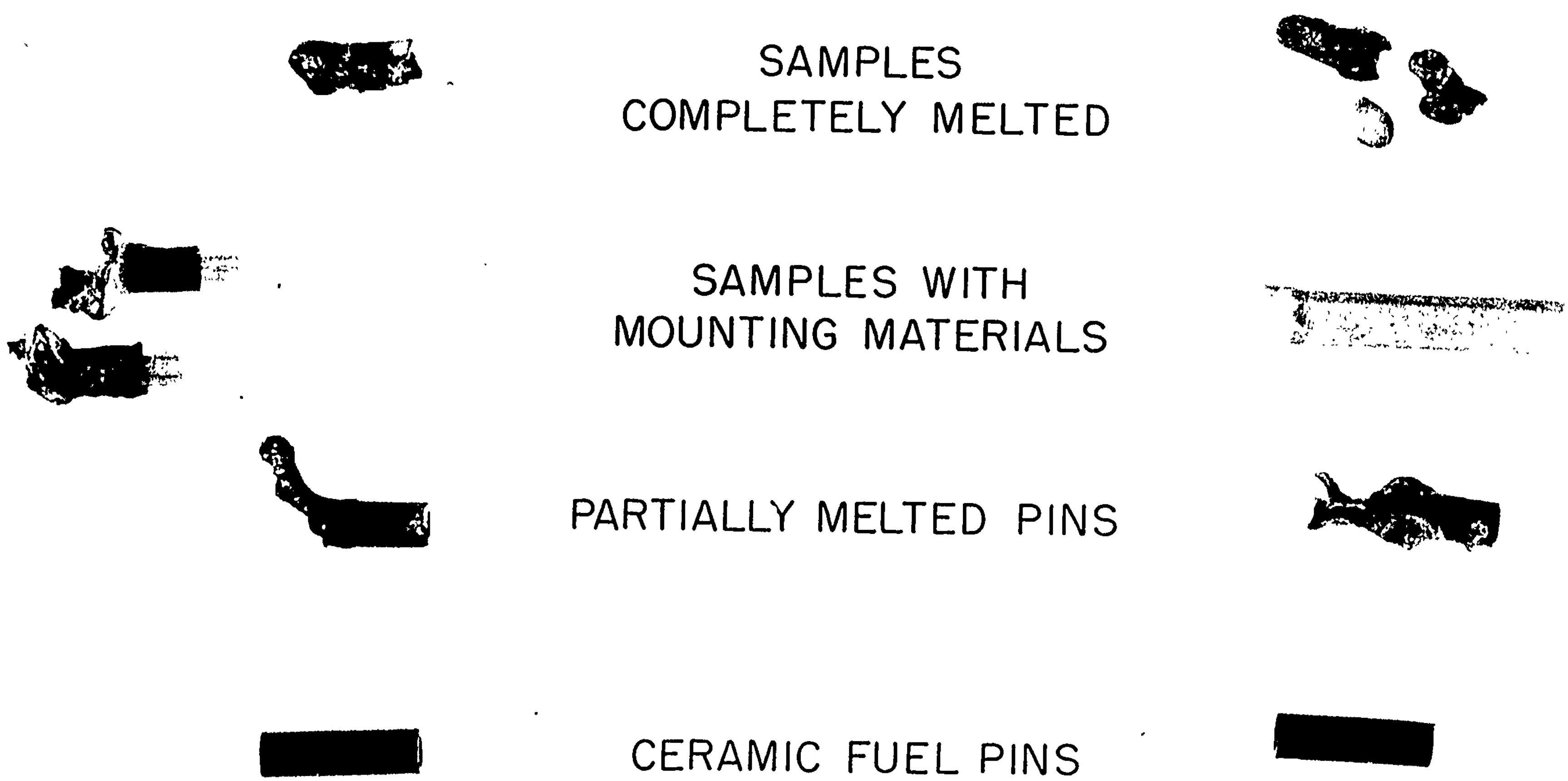

Figure 6
TYPICAL SPECIMENS OF TYPE 14 FUEL

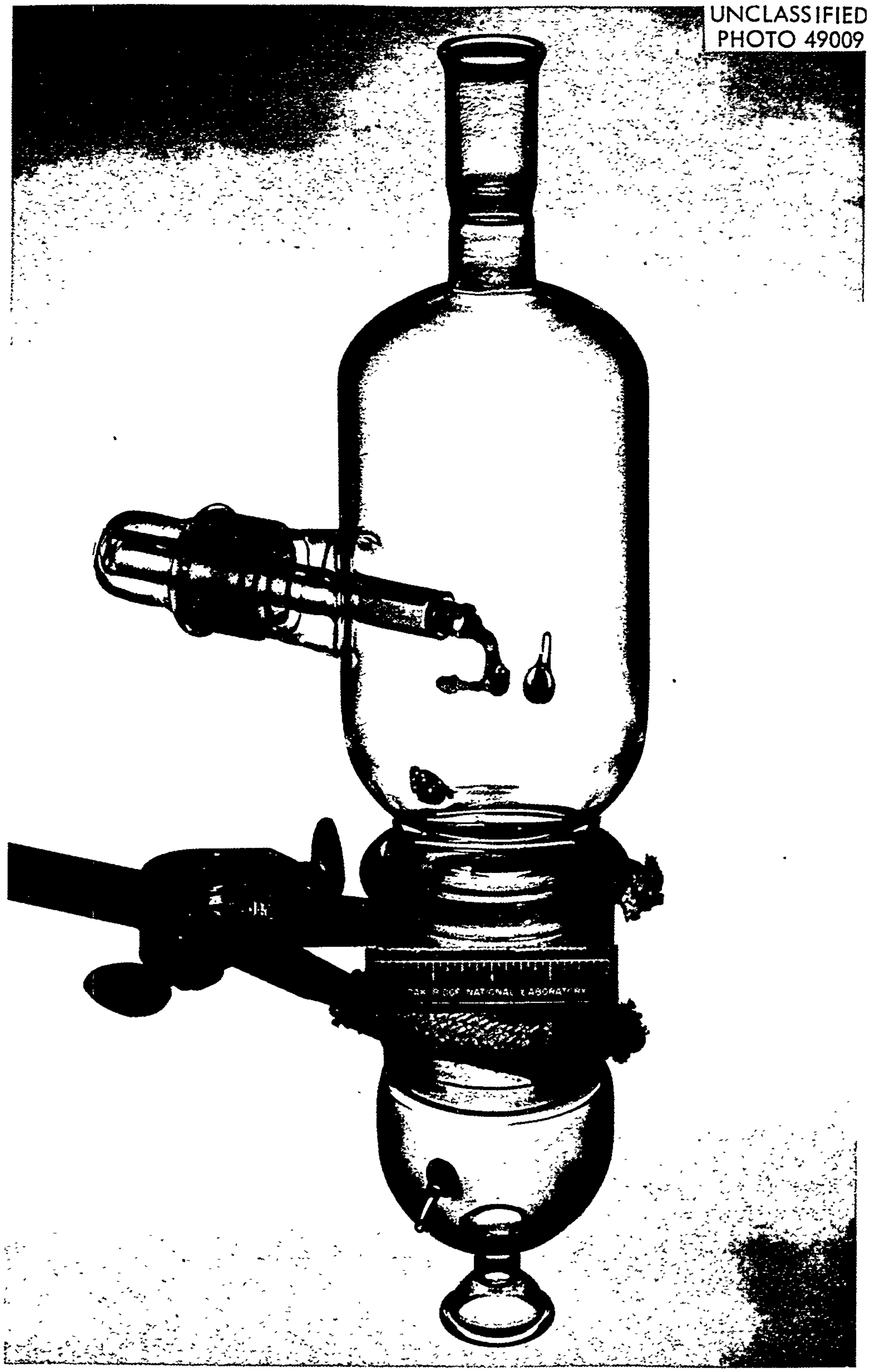

Figure 7

CERAMIC SPECIMEN SHOWN IN CLEAN FURNACE TUBE TO ILLUSTRATE DEFLECTION OF MELTED PORTION FROM HIGH TEMPERATURE ZONE

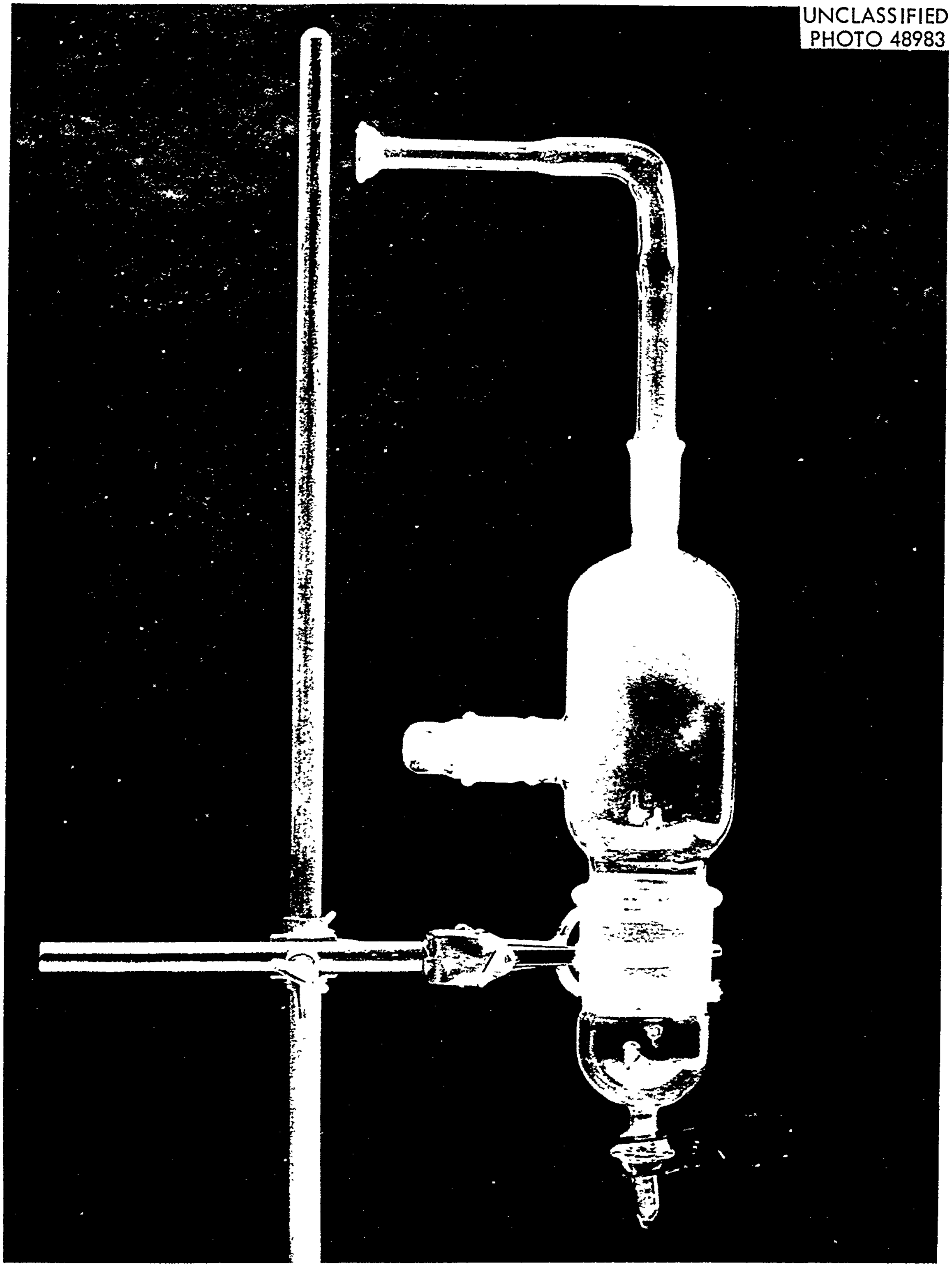

Figure 8

APPEARANCE OF FURNACE TUBE FOLLOWING A MELT WITH OXIDE COATING PLATED ON GLASS

TABLE I.

SUMMARY FOR TYPE 14 GE-ANP F

Run No.	Location In Train	Gross γ Activity	Rare Gas γ	I γ	Te β⁻	Cs γ	Ru γ
	Furnace	9.8		49.3	57.7	56.0	53.9
	Filters	0.7		3.2	5.0	5.5	1.9
1	Charcoal Traps	0.1	75.7	3.7			
	Total	10.6	75.7	56.2	62.7	61.5	
	Furnace	9.5		46.4	62.1	50.7	60.1
	Filters	1.1		10.2	6.1	11.0	3.6
2	Charcoal Traps	0.4	71.3	16.0			
	Total	11.0	71.3	72.6	68.2	62.0	
	Furnace	10.1		62.3	66.8	34.5	63.6
	Filters	1.2		5.4	8.3	15.6	0.8
3	Charcoal Traps	0.4	77.1	11.3			
	Total	11.6	77.1	79.0	75.1	50.1	
Average % Release		11.1	73.7	69.3	68.0	57.9	

* Airflow Across Sample = 0.3 ft./min.

Melting Time: 80 sec. Run - 1, 90 sec. Runs 2 and 3.
Fuel Previously Irradiated to 0.43 % Burn-Up. Re-irradiated 8 hrs. at 7×10^{12} r
Runs 1-2 and 3: Our Ref. No. 1-13-1, 1-14-1 and 1-19-1.

** TRE = Total Rare Earths, Includes Cerium.

L MELTING EXPERIMENTS*

ERCENT OF TOTAL RELEASED ON MELTING

	Ce β^-	TRE** β	Sr β^-	Ba β^-	Zr γ	(UO_2 + UO_3)	Be as (BeO)
	0.53	0.23	0.026	0.3	0.02	7.9	0.9
	0.03	0.02	0.004	0.006		0.9	0.05
.8	0.56	0.25	0.03	0.3	0.02	8.8	0.95
	0.40	0.185	0.14	0.53	0.01	6.7	0.5
	0.06	0.003	0.01	0.01		1.5	0.1
.7	0.46	0.19	0.15	0.54	0.01	8.2	0.6
	0.24	0.26	0.1	0.55	0.02	6.4	1.5
	0.11	0.09	0.01	0.01	0.01	1.4	0.1
.4	0.35	0.35	0.11	0.56	0.03	7.8	1.6
.3	0.46	0.30	0.10	0.47	0.02	8.2	1.05

cm^2/sec. Cooling times 27, 28 and 33 days.

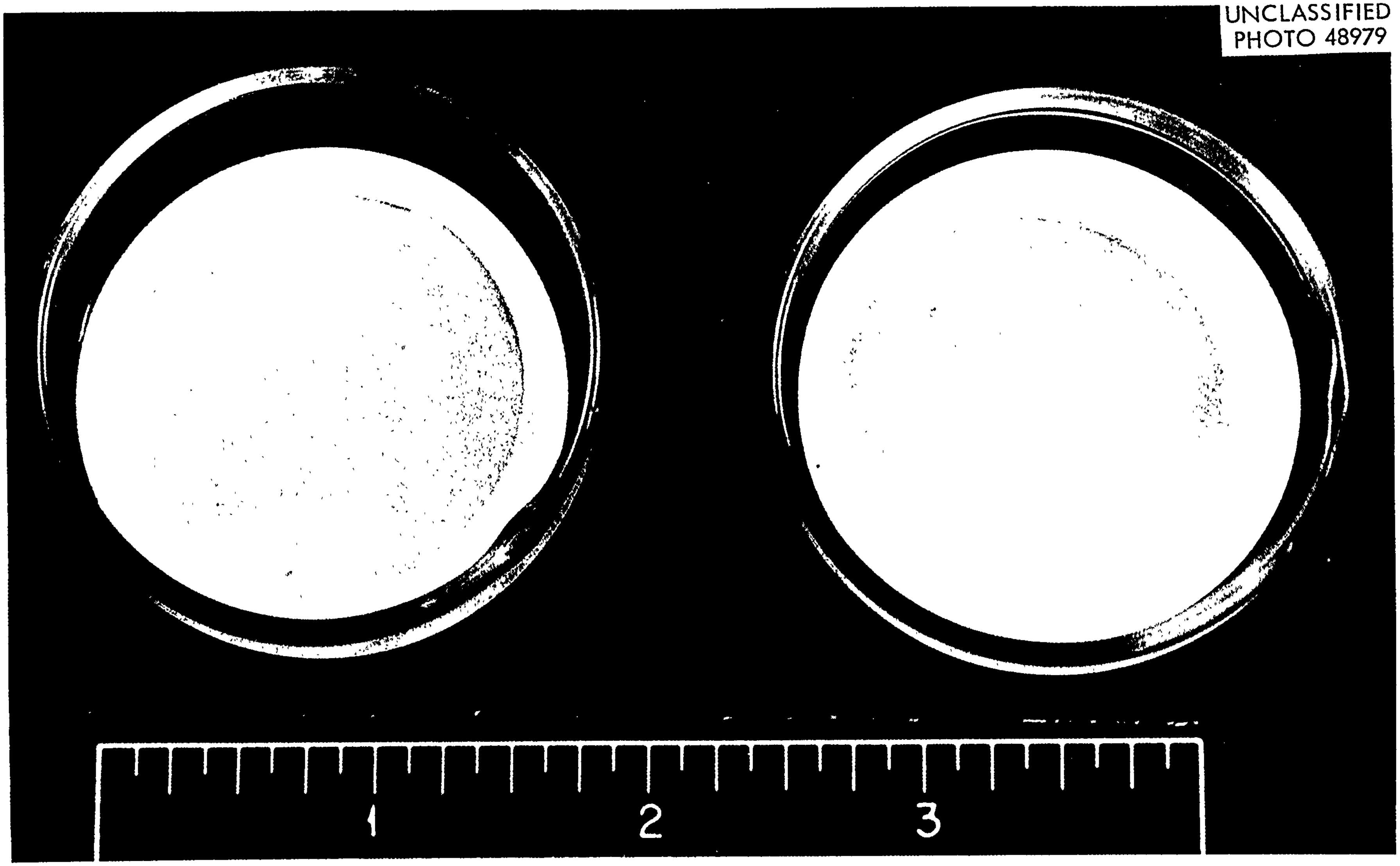

Figure 9

FILTERS FROM RUN ANP-1; DARK SOLIDS SHOWN ON BOTH FILTERS CAUSED BY A TEAR IN THE PAPER

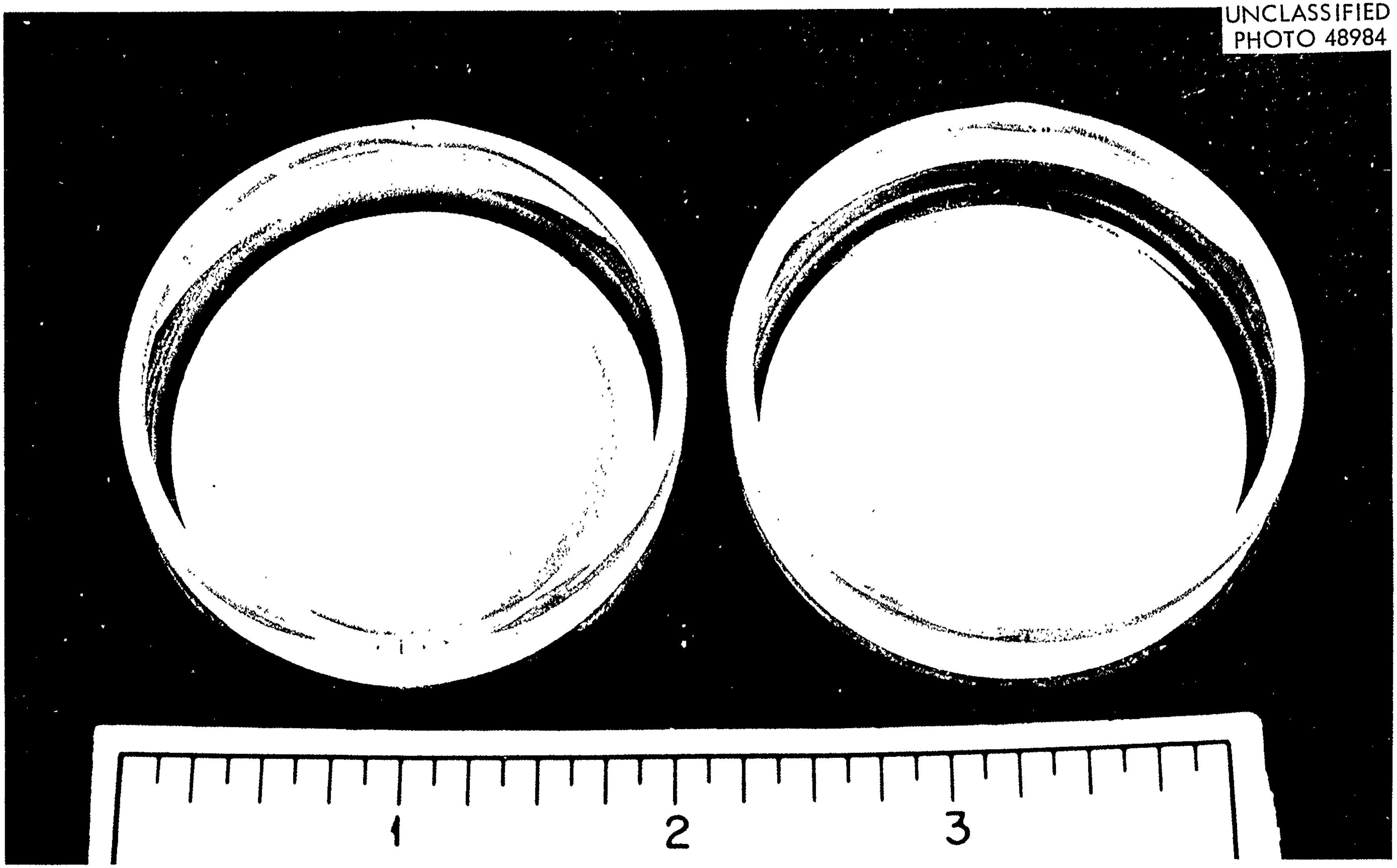

Figure 10

FILTERS FROM ANP RUN NO. 2 SHOWING FIRST FILTER REMAINED INTACT show spherical particles below 0.1 micron.

8.0 Effect of Air Velocity on Plating Out

Logically the plating out process should require some minimum contact time with the furnace wall. Since the initial runs were concluded with a very low air flow (0.5 changes per minute in the furnace tube) a second series was started in which this was raised to 5 changes per minute, or 3.0 linear feet/min. across the sample. The effect of increased velocity was pronounced in that practically all the solids were transferred to the filters and little plate-out occurred. See Table II. The deposition of the large majority of all condensible fission products in the plated out oxides is significant. Only a few percent of the iodine was not contained in this coating.

9.0 Behavior of Individual Fission Products

The results of the first six runs are summarized in Tables I and III. The generally high values for the volatile components (boiling points far below the fuel melting temperature) e.g. xenon and krypton, and elemental iodine, tellurium, cesium and ruthenium (as RuO_4) are not surprising in view of previous results at 1500-1800° C on metallic fuels. The fact that the rare gases are not quantitatively released is anomolous. An attempt is made to explain this on the basis of incomplete melting expecially at the rear of the sample. A special analysis of this region is being conducted. Another possible explanation is that no soluble gases are generated and released in the melting to produce a sweeping action which is normally required for complete release.

Both a disadvantage and an advantage over metallic fuels is illustrated here. In many metallic systems both tellurium and ruthenium are contained by remaining alloyed in the unoxidized metal. Such is not the case here; however, the refractory oxides of Ba, Sr, Zr, rare-earths, etc. are most completely retained.

10.0 Effect of Burn-Up

Only one degree of burn-up is represented in the LTC-14 type samples used. These had 0.43% burn-up and a cooling period of 1 1/2 years. A significant difference is noted in two samples No. 4, and 5 (Table II) which were irradiated to only a trace level (~ 0.01% burn-up), particularly in the cesium release. A general effect is noted in most of the other fission products but none to the extent evident in cesium. At higher burn-up ratios, a more pronounced effect should appear.

11.0 Extension of the ANP Program

In addition to temperature measurment and a time-at-temperature effect mentioned above; additional samples of types LTC 25 and 26 which are protectively coated with BeO and ZrO_2 will be melted. These have a somewhat heavier wall thickness which may add

TABL

SUMMARY FOR TYPE 14 FU

Run No.**	Location in Train	Gross γ Activity	Rare Gas γ	I γ	Te β	Cs γ	Ru γ
	Furnace	8.8		12.1	28.6	18.3	33.0
6	Filters	6.2		9.8	43.2	30.9	24.6
	Charcoal Traps	3.4	69.5	56.9			
Total % Released		18.4***	69.5	78.8	71.8	49.2	5

*Air Flow = 3000 cc/min. or 5 linear feet/min. across sample.

**Same as Ref. No. 1-26-1.

Sample reirradiated 168 hrs. at 10^{12} n/Cm^2/sec. Cooling time, 8 days.

Previous burnup 0.43%, Type 14 fuel.

***High value dependent upon relatively shorter cooling time.

II

WITH HIGH AIR FLOW.*

	PERCENT OF TOTAL RELEASED ON MELTING						
	Ce β^-	TRE** β	Sr β^-	Ba β^-	Zr γ	(UO_2 + UO_3)	Be as (BeO)
	0.17	0.1	0.05	0.08	0.02	4.1	0.53
	0.37	0.18		0.07	0.01	6.9	0.10
6	0.54	0.28	0.05	0.15	0.03	11.0	0.63

TA

SUMMARY FOR TRACE I

Run No.	**Location In Train	Gross γ Activity	Rare Gas γ	I γ	Te β^-	Cs γ	Ru γ
	Furnace	6.62		50.9	66.2	40.0	53.9
4	Filters	0.66		10.7	0.1	5.7	0.3
	Charcoal Traps	0.20	73.8	9.1			
	Total	7.48	73.8	70.7	66.3	45.7	
	Furnace	8.66		58.3	72.0	31.3	57.3
5	Filters	1.06		12.0	12.5	10.1	4.6
	Charcoal Traps	0.29	82.0	13.2			
	Total	10.0	82.0	83.5	84.5	41.4	
Average % Released		8.74	77.9	77.1	75.4	43.6	

* Irradiation: 8 hrs. at 7 x 10^{12} n/Cm^2/sec.: Cooling Time 35 and 36 days.

** Run 4, Ref. No. 1-21-1, Run 5, Ref. No. 1-22-1.

Total Heating Time 90 sec.

Air Flow Run 4, 0.3 l.ft./min., Run 5, 0.4 l.ft./min.

E III

ADIATED TYPE 14 FUEL*

PERCENT OF TOTAL RELEASED ON MELTING

	Ce β^-	TRE** β	Sr β^-	Ba β^-	Zr γ	(UO_2 + UO_3)	Be as (BeO)
	0.40	0.06	0.015	0.17	0.42	8.88	1.2
	0.10	0.09	0.002	0.05		0.4	0.04
		0.03					
.2	0.50	0.18	0.17	0.22	0.42	9.3	1.24
	0.29	0.08	0.12	0.1	0.01	9.4	0.52
	0.10	0.03		00.03		2.0	0.04
.9	0.39	0.11	0.12	0.13	0.01	11.4	0.56
.0	0.44	0.15	0.15	0.18	0.21	10.4	0.90

somewhat to the diffusion time effect. In spite of this with approximately the same degree of burn-up, no large difference in release behavior is expected.

12.0 Discussion of Results

In the results to date, while unfortunately high, there is an unusual degree of consistency in the release values. The tables have been prepared to reveal as much detail as possible in the plate-out phenomena including the effect of increasing air flow to the point that most of the vaporized solids are transferred to the filters. At the same time a larger portion of the iodine is carried through to the charcoal absorbers. It is somewhat significant that the observed releases for trace level fission products are only slightly different from those found with 0.43% burn-up. This effect is much more pronounced in metallic systems.

It may be obvious that in a thin tubular fuel sample where diffusion lengths are minimized and especially at these extreme temperatures, fission products of lower boiling points should be volatilized rapidly. Such is the case except for those forming refractory oxides e.g., Sr, Ba, Zr, and rare earths, which are apparently retained in solid solution with BeO.

13.0 References

1. A. D. Little, Co., Acorn Park, Cambridge, Massachusetts.

2. M. R. Null and W. W. Lozier, "Carbon Arc-Image Furnaces," Review of Sci. Instruments, 29, No. 2, 163-70, Feb. 1958.